

(12) PATENT
(19) AUSTRALIAN PATENT OFFICE

(11) Application No. **AU 199728540 B2**
(10) Patent No. **728167**

(54) Title
Process for the removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas and system therefor

(51)⁷ International Patent Classification(s)
B01D 053/62

(21) Application No: **199728540**

(22) Application Date: **1997.07.10**

(30) Priority Data

(31) Number	(32) Date	(33) Country
8-226543	1996.08.28	JP

(43) Publication Date : **1998.03.05**

(43) Publication Journal Date : **1998.03.05**

(44) Accepted Journal Date : **2001.01.04**

(71) Applicant(s)
Mitsubishi Heavy Industries, Ltd

(72) Inventor(s)
Masaki Iijima; Shigeaki Mitsuoka; Hiroshi Tanaka

(74) Agent/Attorney
LORD and COMPANY, 4 Douro Place, WEST PERTH WA 6005

(56) Related Art
EP 768365

ABSTRACT

This invention provides a process for the removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas wherein a raw gas (1) containing carbon dioxide is
5 fed to a decarbonation tower (2) where the raw gas (1) is brought into gas-liquid contact with a partially regenerated absorbing fluid (21) in a lower absorption section (3) so as to cause carbon dioxide to be partially absorbed thereinto, and then brought into gas-liquid contact with a regenerated
10 absorbing fluid (22) in an upper absorption section (4) so as to cause carbon dioxide to be absorbed thereinto until a very low carbon dioxide concentration is reached; and high-pressure carbon dioxide is recovered from the resulting carbon dioxide-loaded absorbing fluid (20), as well as a
15 system therefor.

This invention makes it possible to remove highly concentrated carbon dioxide from high-pressure natural gas and various synthesis gases with the aid of a carbon dioxide absorbing fluid until a very low carbon dioxide concentration
20 is reached and, moreover, to recover high-pressure carbon dioxide from the absorbing fluid.

AUSTRALIA
PATENTS ACT 1990
COMPLETE SPECIFICATION
FOR A STANDARD PATENT
(Original)

APPLICATION NO:
LODGED:

COMPLETE SPECIFICATION LODGED:
ACCEPTED:
PUBLISHED:

RELATED ART:

NAME OF APPLICANT:

MITSUBISHI HEAVY INDUSTRIES,
LTD

ACTUAL INVENTORS:

MASAKI IJIMA ;
SHIGEAKI MITSUOKA; and
HIROSHI TANAKA

ADDRESS FOR SERVICE:

LORD & COMPANY,
Patent & Trade Mark Attorneys,
of 4 Douro Place, West Perth,
Western Australia, 6005, AUSTRALIA.

INVENTION TITLE:

"PROCESS FOR THE REMOVAL
AND HIGH-PRESSURE RECOVERY
OF CARBON DIOXIDE FROM A
HIGH-PRESSURE RAW GAS AND
SYSTEM THEREFOR"

The following Statement is a full description of this invention including the best method of performing it known to me:

SPECIFICATION

TITLE OF THE INVENTION

Process for the Removal and High-Pressure Recovery of
Carbon Dioxide from a High-Pressure Raw Gas and System
Therefor

BACKGROUND OF THE INVENTION

1. Field of the invention

This invention relates to a process for treating a high-pressure raw gas selected from high-pressure natural gas and various synthesis gases with a carbon dioxide absorbing fluid, whereby highly concentrated carbon dioxide (CO₂) is fully removed from the raw gas to obtain a refined gas having a carbon dioxide concentration of 10 to 10,000 ppm and, moreover, for recovering high-pressure carbon dioxide from the absorbing fluid, as well as a system therefor.

2. Description of the related art

Usually, high-pressure natural gas may contain a considerable amount (e.g., 10 to 20%) of carbon dioxide. When such natural gas is converted to liquefied natural gas (hereinafter referred to as LNG), carbon dioxide forms dry ice as a result of cooling, and this solid material may cause problems such as obstruction of the system. Accordingly, its carbon dioxide concentration must be reduced to 50 ppm or less.

Moreover, in the case of ammonia-urea synthesis, a gaseous

1 mixture composed of hydrogen, carbon monoxide, carbon dioxide
and the like is obtained by the partial oxidation or steam
reforming of natural gas, naphtha or the like. This gaseous
mixture is subjected to a CO shift reaction for converting
5 carbon monoxide to carbon dioxide, and used as a raw material
for ammonia synthesis after the separation of carbon dioxide.
On the other hand, the separated carbon dioxide is
pressurized to 100-200 atmospheres and reacted with ammonia
to form urea. Accordingly, the carbon dioxide concentration
10 of a raw material for ammonia synthesis must generally be
reduced to the order of 500 ppm.

Furthermore, in the case of hydrogen gas for use in
chemical syntheses, its carbon dioxide concentration must be
reduced to a level ranging from several tens to several
15 thousands of parts per million, depending on its application.

In order to use the separated carbon dioxide for the
purpose of tertiary oil recovery (EOR), liquid carbon dioxide
production or urea synthesis or store it in an underground
aquifer as a countermeasure against global warming, it must
20 be pressurized to a pressure ranging from several tens to
several hundreds of atmospheres.

Consequently, it would be desirable to remove carbon
dioxide from an inherently high-pressure raw gas until a very
low carbon dioxide concentration is reached and recover the
25 removed carbon dioxide in a high-pressure state. However, no

method useful for this purpose has been known in the prior art.

In the prior art, the following process has been employed to remove carbon dioxide until a very low carbon dioxide concentration (for example, of the order of 100 ppm) is reached. First of all, a gas is fed to the bottom of a decarbonation tower while a carbon dioxide absorbing fluid is fed to the top of the decarbonation tower. Thus, the gas is brought into gas-liquid contact with the absorbing fluid, so that carbon dioxide is absorbed thereinto and removed. Then, the absorbing fluid having carbon dioxide absorbed thereinto (hereinafter referred to as the loaded absorbing fluid) is introduced into a regeneration tower where carbon dioxide is liberated by steam stripping to obtain a regenerated absorbing fluid. This regenerated absorbing fluid is fed to the decarbonation tower.

Moreover, another example of the method for removing carbon dioxide from high-pressure natural gas and recovering the removed carbon dioxide in a high-pressure state is a process for the bulk removal of carbon dioxide.

However, the above-described conventional processes for the high removal and high-pressure recovery of carbon dioxide and systems therefor involve the following problems.

(1) In the above-described process using a carbon dioxide absorbing fluid, the carbon dioxide concentration of the

regenerated absorbing fluid can be reduced and, therefore, carbon dioxide can be removed until a very low carbon dioxide concentration is reached. However, since the absorbing fluid is regenerated by depressurization, it is difficult to
5 recover carbon dioxide having a sufficiently high pressure. Consequently, it is necessary to pressurize the recovered carbon dioxide again by means of a compressor.

(2) In the above-described process for the bulk removal of carbon dioxide, the natural gas from which carbon dioxide has been removed still contains about 1 to 5% of carbon dioxide.

SUMMARY OF THE INVENTION

The present invention has been made in view of the above-described existing state of the art, and it is an object thereof to provide a process for removing highly concentrated carbon dioxide from high-pressure natural gas and various
15 synthesis gases with the aid of a carbon dioxide absorbing fluid until a very low carbon dioxide concentration is reached and, moreover, for recovering high-pressure carbon dioxide from the absorbing fluid.

20 As a result of intensive investigations, the present inventors have found that carbon dioxide can be highly removed from a raw gas and, at the same time, carbon dioxide can be recovered in a high-pressure state, by treating the raw gas with an absorbing fluid so as to cause carbon dioxide
25 to be absorbed therein, heating the resulting carbon

dioxide-loaded absorbing fluid to liberate carbon dioxide in a high-pressure state and thereby regenerate the absorbing fluid partially, recycling a portion of the partially regenerated absorbing fluid to the absorption step, and
5 highly regenerating the remainder of the partially regenerated absorbing fluid and recycling the resulting highly regenerated absorbing fluid to the absorption step. The present invention has been completed on the basis of this finding.

10 That is, the present invention provides a process for the high removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas which comprises the steps of feeding a high-pressure raw gas containing 2 to 50% of carbon dioxide and having a pressure of not less than 2 kg/cm²
15 (absolute pressure) to a decarbonation tower consisting of a lower absorption section and an upper absorption section, where the raw gas is brought into gas-liquid contact with a partially regenerated absorbing fluid in the lower absorption section so as to cause carbon dioxide to be partially
20 absorbed thereinto, and then brought into gas-liquid contact with a regenerated absorbing fluid in the upper absorption section so as to cause carbon dioxide to be absorbed thereinto and removed until a very low carbon dioxide concentration is reached, and the refined gas having a carbon
25 dioxide concentration of 10 to 10,000 ppm is discharged out

of the system; heating the carbon dioxide-loaded absorbing fluid produced in the decarbonation tower and feeding it to a high-pressure regeneration tower where some carbon dioxide is liberated under a pressure ranging from 2 kg/cm² (absolute pressure) to the pressure of the raw gas to obtain a partially regenerated absorbing fluid, and a portion of the partially regenerated absorbing fluid is fed to the lower absorption section; feeding the remainder of the partially regenerated absorbing fluid to a low-pressure regeneration tower where carbon dioxide is liberated to obtain a regenerated absorbing fluid, and the regenerated absorbing fluid is fed to the upper absorption section; recovering high-pressure carbon dioxide by cooling the carbon dioxide liberated under pressure in the high-pressure regeneration tower and separating it from any entrained water; and recovering carbon dioxide by cooling the carbon dioxide liberated in the low-pressure regeneration tower and separating it from any entrained water, as well as a system therefor.

The present invention makes it possible to remove highly concentrated carbon dioxide from high-pressure natural gas and various synthesis gases until a very low carbon dioxide concentration of not greater than 1,000 ppm is reached and, moreover, to recover high-pressure carbon dioxide. As a result, the compression power required for subsequent use of

the carbon dioxide can be saved and the scale of the equipment can be reduced.

BRIEF DESCRIPTION OF THE DRAWINGS

FIG. 1 is a flow diagram illustrating the process for the high removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas in accordance with the present invention; and

FIG. 2 is a flow diagram illustrating a conventional process for the high removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas.

DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

The raw gas treated according to the present invention is selected from natural gas and various synthesis gases which have a high pressure and contain carbon dioxide. Examples of such gases are carbon dioxide-containing gases which are obtained by the steam reforming or partial oxidation of naphtha, natural gas, heavy oil, coke or the like and have been subjected to a CO shift reaction for converting carbon monoxide to carbon dioxide. Preferably, these gases are used in the present invention after having been desulfurized to a sulfur compound concentration of not greater than 100 ppm.

Although no particular limitation is placed on the pressure of the raw gas, it is preferably not less than 2 kg/cm² and more preferably not less than 10 kg/cm². The upper limit of the pressure is not specifically defined, but

is usually in the range of 200 to 300 kg/cm². Similarly, although no particular limitation is placed on the carbon dioxide concentration of the raw gas, it is preferably in the range of 1 to 50% by volume and more preferably 10 to 30% by volume.

After being treated according to the present invention, the resulting purified gas has substantially the same pressure as the raw gas and a carbon dioxide concentration of 1 to 10,000 ppm and preferably 10 to 1,000 ppm.

The carbon dioxide absorbing fluid used in the present invention comprises an aqueous solution containing an absorbent selected from various basic compounds and mixtures thereof.

For this purpose, it is preferable to select an absorbing fluid having the following absorption capacity.

After having absorbed carbon dioxide by gas-liquid contact with a raw gas, the carbon dioxide absorbing fluid is partially regenerated in a high-pressure regeneration tower to liberate some of the carbon dioxide therefrom, and then recycled for use in the lower absorption step. Thus, its absorption capacity must be such that, when the partial pressure of carbon dioxide is 2 kg/cm² or greater, the difference in saturated carbon dioxide absorption level between the absorption temperature (e.g., 40°C) and the partial regeneration temperature (e.g., 120°C) is not less

than a specific value (e.g., not less than 30 Nm³ of CO₂ per m³ of the absorbing fluid and preferably not less than 40 Nm³ of CO₂ per m³ of the absorbing fluid).

5 Usually, if the temperature and the partial pressure of carbon dioxide are given, the saturated carbon dioxide absorption level of the aforesaid absorbing fluid shows a fixed value based on the saturated carbon dioxide absorption curve for that absorbing fluid, almost regardless of the type of the carbon dioxide-containing gas.

10 In the present invention, as shown in the Examples which will be given later, carbon dioxide is removed from high-pressure natural gas having a pressure of 30 kg/cm² or greater by absorbing it into a partially regenerated absorbing fluid, and the resulting loaded absorbing fluid is transferred to a partial regeneration step where, without
15 being depressurized, it is heated to liberate carbon dioxide therefrom. Accordingly, it is preferable to use an absorbing fluid which can easily absorb carbon dioxide at a low
20 temperature and a low partial pressure in the absorption step and can easily liberate carbon dioxide at a high temperature and a high partial pressure in the regeneration step.

Consequently, a physical absorbing fluid or an absorbing fluid comprising amine having strong physical absorption properties is preferably used as the aforesaid carbon dioxide
25 absorbing fluid.

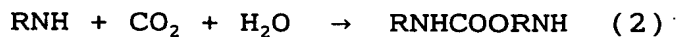
The physical absorbing fluid includes methanol and polyethylene glycol dimethyl ether.

As for the amine having strong physical absorption properties, specifically, amines, amino acids and alkali metal salts of the amino acids are used. If necessary, amines having strong chemical absorption properties, alkali metal carbonates and the like may be added thereto.

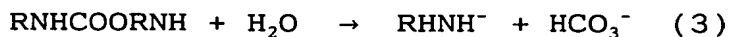
Typical examples of the amines having strong physical absorption properties are tertiary amines, which mainly react below.



Hence, the tertiary amines may be easily regenerated by flash or the like. On the other hand, primary amines and secondary amines are bases having strong chemical absorption properties and mainly react below.



The regeneration requires heat of decomposition. Even in the case of the equation (2), however, the amines with high steric hindrance tend to form a bond having closer characteristic to an ionic bond than a chemical bond, resulting the further reaction below.



Accordingly, the amines having strong physical absorption properties include not only tertiary amines but also primary amines and secondary amines.

Hence, preferred examples of the amines are hindered amines. The preferred hindered amines include N-methyldiethanolamine (MDEA), triethanolamine (TEA), dimethylamino-1,3-propanediol (DMAPD) and diethylamino-1,3-propanediol (DEAPD).

Useful alkali metal salts include potassium carbonate.

Moreover, carbon dioxide absorption promoters such as piperazine, substituted piperazines, piperidine and substituted piperidines may be added to the above-described carbon dioxide absorbing fluid.

The carbon dioxide absorbing fluid may be a 20-80% aqueous solution of the absorbent, though the concentration depends on the type of the absorbent and the conditions of use.

If necessary, a solvent such as N-methylpyrrolidone or sulfolane may be added to the carbon dioxide absorbing fluid.

The high-pressure carbon dioxide recovered from the carbon dioxide absorbing fluid has a pressure of not less than 2 kg/cm² and preferably a pressure ranging from 10 kg/cm² to a value almost equal to the pressure of the raw gas, though the pressure depends on the type of the raw gas and the use of carbon dioxide.

Accordingly, the high-pressure regeneration tower is operated at the aforesaid pressure, and the loaded absorbing fluid is heated to a temperature of 90 to 150°C and preferably 100 to 140°C. Thus, water and carbon dioxide are

partially liberated from the loaded absorbing fluid, whereby the loaded absorbing fluid is partially regenerated.

The partially regenerated absorbing fluid is more highly regenerated under conditions including an operating pressure of not greater than 2 kg/cm² and preferably 0.5 to 1 kg/cm² and a fluid temperature of 100 to 150°C and preferably 110 to 140°C, for example, by circulating it through a reboiler and heating it therewith.

The degree of regeneration of the partially regenerated absorbing fluid fed to the lower absorption section of the decarbonation tower, the degree of regeneration of the regenerated absorbing fluid fed to the upper absorption section thereof, and the feed ratio of them depend on the type of the absorbing fluid, the carbon dioxide concentration, pressure and other conditions of the raw gas, the carbon dioxide concentration of the refined gas, and the flow rate, concentration, pressure and other conditions of the recovered high-pressure carbon dioxide. For example, the degree of partial regeneration is in the range of 0.05 to 0.3 mole of CO₂ per mole of the absorbing fluid, the degree of regeneration is in the range of about 0.01 to 0.1 mole of CO₂ per mole of the absorbing fluid, and the proportion of the partially regenerated absorbing fluid fed to the decarbonation tower is in the range of 30 to 95%.

The process of the present invention is specifically

described below with reference to FIG. 1.

A raw gas 1 is fed to the bottom of a decarbonation tower 2. In a lower absorption section 3, raw gas 1 comes into gas-liquid contact with a partially regenerated absorbing fluid 21, so that carbon dioxide is partially absorbed thereinto. Then, in an upper absorption section 4, raw gas 1 comes into gas-liquid contact with a regenerated absorbing fluid 22, so that carbon dioxide is further absorbed until a very low carbon dioxide concentration is reached. The resulting gas is discharged out of the system as a refined gas 17.

In the above-described process, regenerated absorbing fluid 22 flows down through upper absorption section 4 while coming into gas-liquid contact with raw gas 1, and mixes with partially regenerated absorbing fluid 21 in lower absorption section 3. The resulting absorbing fluid flows down through lower absorption section 3 while coming into gas-liquid contact with raw gas 1, and is withdrawn from the bottom of decarbonation tower 2.

The carbon dioxide-loaded absorbing fluid 20 emerging from the bottom of decarbonation tower 2 is heated, if necessary, by heat exchange with partially regenerated absorbing fluid 21 or regenerated absorbing fluid 22 in a heat exchanger 5 or 6, further heated to a predetermined temperature by means of a heater 7, and then fed to a high-temperature regeneration

tower 8.

The resulting partially regenerated absorbing fluid 21 is withdrawn from the bottom of high-temperature regeneration tower 8. A portion thereof is cooled in heat exchanger 5, further cooled in heat exchanger 15 if necessary, and then fed to lower absorption section 3.

The mixture of water and carbon dioxide discharged from the top of high-pressure regeneration tower 8 is cooled in a condenser 9 and introduced into a gas-liquid separator 10 where it is separated into water and high-pressure carbon dioxide 18. The water is recycled to the top of high-pressure regeneration tower 8. If necessary, some of the water may be fed to the top of absorption tower 2, or used for recovery of the absorbing fluid or as recycle water to a low-pressure regeneration tower.

The remainder of partially regenerated absorbing fluid 21 is fed to a low-pressure regeneration tower 11 which is operated at an absolute pressure of less than 2 kg/cm² or at reduced pressure and equipped with a reboiler 12 to heat the bottom fluid. Thus, water and carbon dioxide are liberated from the partially regenerated absorbing fluid, so that it is highly regenerated. The degree of regeneration of the loaded absorbing fluid is determined by the heating temperature, residence time and operating pressure employed for the treatment of the loaded absorbing fluid in low-pressure

regeneration tower 11.

The regenerated absorbing fluid 22 is withdrawn from the bottom of low-pressure regeneration tower 11. A portion thereof is cooled in heat exchanger 6, further cooled in heat exchanger 16 as required, and then fed to upper absorption section 4.

The mixture of water and carbon dioxide discharged from the top of low-pressure regeneration tower 11 is cooled in a condenser 13 and introduced into a gas-liquid separator 14 where it is separated into water and low-pressure carbon dioxide 19. The water is recycled to the top of low-pressure regeneration tower 11. If necessary, some of the water may be fed to the top of absorption tower 2, or used for recovery of the absorbing fluid or as recycle water to the high-pressure regeneration tower.

The absorption tower and the several regeneration towers may comprise plate towers or packed towers, provided that they can bring about efficient gas-liquid contact and cause a small pressure loss. To this end, there may be used any of various conventional packing materials such as those of the wetted-wall type.

Greater economy is achieved as the proportion of high-pressure carbon dioxide recovered according to the present invention becomes higher. In the present invention, not less than 50%, preferably not less than 70%, and most preferably

not less than 90% of carbon dioxide can be recovered at high pressure.

Thus, the present invention makes it possible to recover carbon dioxide at high pressure, resulting in a great saving of compression power and a reduction in the scale of the equipment.

The present invention is further illustrated by the following examples. However, these examples are not to be construed to limit the scope of the invention.

Example 1

This example relates to an application of the process illustrated in the flow diagram of FIG. 1 in which a raw gas for use in ammonia synthesis obtained by the reforming of natural gas was treated with an absorbing fluid comprising an aqueous solution containing 45% by weight of MDEA and 4% by weight of piperazine to remove carbon dioxide therefrom.

A raw gas comprising hydrogen, nitrogen, hydrocarbons and 19% by volume of carbon dioxide was fed to the lower part of an absorption tower under conditions including a pressure of 34 kgG/cm², a temperature of 45°C and a flow rate of 292,000 Nm³/hr.

In a lower absorption section of the absorption tower, the ascending raw gas came into gas-liquid contact with 1,716 m³/hr of a partially regenerated absorbing fluid. Thus, in the lower absorption section, carbon dioxide was partially

removed from the raw gas until its carbon dioxide concentration was reduced to about 2% by volume. Moreover, the raw gas came into gas-liquid contact with 163 m³/hr of a regenerated absorbing fluid in an upper absorption section.

5 The refined raw gas, which had a carbon dioxide concentration of 490 ppm, a temperature of 39°C and a pressure of 33 kgG/cm², was discharged from the top of the absorption tower.

The loaded absorbing fluid having carbon dioxide absorbed thereinto had a fluid temperature of 55°C and contained about 78 Nm³ of carbon dioxide (CO₂) per m³ of the absorbing fluid. This loaded absorbing fluid was subjected to heat exchange, then heated to 120°C, and fed to a high-pressure regeneration tower where it was partially regenerated. The liberated carbon dioxide and water, together with a small amount of the absorbing fluid, were cooled in a condenser and introduced into a gas-liquid separator where carbon dioxide was separated. The cooled and separated carbon dioxide had a temperature of about 40°C, a pressure of 10 kgG/cm² and a flow rate of 49,800 Nm³/hr. This carbon dioxide was 20 pressurized to 190 kgG/cm² by means of a compressor (not shown) and used for the synthesis of urea.

On the other hand, the partially regenerated absorbing fluid withdrawn from the bottom of the high-pressure regeneration tower had a temperature of 100°C and contained 25 about 43 Nm³ of carbon dioxide (CO₂) per m³ of the absorbing

fluid. A portion of this partially regenerated absorbing fluid was subjected to heat exchange with the loaded absorbing fluid, further cooled to 38°C, and fed to the lower absorption section of the absorption tower.

5 The remainder (163 m³/hr) of the partially regenerated absorbing fluid was fed to a low-pressure regeneration tower where it was depressurized to 0.85 kgG/cm² and regenerated with carbon dioxide and water discharged from the top of the tower. The bottom fluid was heated to about 110°C by means
10 of a reboiler provided at the bottom of the low-pressure regeneration tower, and recycled to the low-pressure regeneration tower. Thus, there was obtained a regenerated absorbing fluid containing 1.2 Nm³ of carbon dioxide (CO₂) per m³ of the absorbing fluid. The liberated carbon dioxide
15 and water, together with a small amount of the absorbing fluid, were cooled in a condenser and introduced into a gas-liquid separator where carbon dioxide was separated. The cooled and separated carbon dioxide had a temperature of about 40°C, a pressure of 0.35 kgG/cm² and a flow rate of
20 5,700 Nm³/hr.

The regenerated absorbing fluid withdrawn from the bottom of the low-pressure regeneration tower had a temperature of about 110°C and was fed to the upper absorption section of the absorption tower. During this course, the regenerated
25 absorbing fluid having a temperature of 110°C was subjected

to heat exchange with the loaded absorbing fluid having a lower temperature, and further cooled to 38°C.

Of all the carbon dioxide recovered in the above-described manner, 90% was recovered in the high-pressure regeneration tower and 10% was recovered in the low-pressure regeneration tower.

Thus, the carbon dioxide concentration of the raw gas for use in ammonia synthesis was reduced to a level sufficient for supply to an ammonia synthesis process. Moreover, the pressure of the recovered carbon dioxide was so high that the compression power required for the synthesis of urea could be saved and the scale of the equipment could be reduced.

Example 2

This example relates to an application of the process illustrated in FIG. 1 in which a raw gas for use in ammonia synthesis obtained by the reforming of natural gas was treated with an absorbing fluid comprising an aqueous solution containing 45% by weight of MDEA and 4% by weight of piperazine to remove carbon dioxide therefrom.

A raw gas 1 comprising hydrogen, nitrogen, hydrocarbons and 19% by volume of carbon dioxide was fed to the lower part of an absorption tower 2 under conditions including a pressure of 34 kgG/cm², a temperature of 45°C and a flow rate of 292,000 Nm³/hr.

In a lower absorption section 3 of absorption tower 2, the

ascending raw gas 1 came into gas-liquid contact with a partially regenerated absorbing fluid 21 ($1,716 \text{ m}^3/\text{hr}$). Thus, in lower absorption section 3, carbon dioxide was partially removed from raw gas 1 until its carbon dioxide concentration was reduced to about 2% by volume. Moreover, raw gas 1 came into gas-liquid contact with a regenerated absorbing fluid 22 ($200 \text{ m}^3/\text{hr}$) in an upper absorption section 4. The refined gas, which had a carbon dioxide concentration of 50 ppm, a temperature of 39°C and a pressure of $33 \text{ kgG}/\text{cm}^2$, was discharged from the top of absorption tower 2.

The loaded absorbing fluid 20 having carbon dioxide absorbed thereinto contained about 78 Nm^3 of carbon dioxide (CO_2) per m^3 of the absorbing fluid. This loaded absorbing fluid 20 was subjected to heat exchange, then heated to 120°C , and fed to a high-pressure regeneration tower 8 where it was partially regenerated. The liberated carbon dioxide and water, together with a small amount of the absorbing fluid, were cooled in a condenser 9 and introduced into a gas-liquid separator 10 where carbon dioxide 18 was separated. The cooled and separated carbon dioxide 18 had a temperature of about 40°C , a pressure of $33 \text{ kgG}/\text{cm}^2$ and a flow rate of $49,640 \text{ Nm}^3/\text{hr}$. This carbon dioxide 18 was pressurized to $150 \text{ kgG}/\text{cm}^2$ by means of a compressor (not shown) and used for the synthesis of urea.

On the other hand, the partially regenerated absorbing

fluid 21 withdrawn from the bottom of high-pressure regeneration tower 8 had a temperature of 100°C and contained 29.2 Nm³ of carbon dioxide (CO₂) per m³ of the absorbing fluid. A portion of this partially regenerated absorbing fluid 21 was subjected to heat exchange with loaded absorbing fluid 20, further cooled to 38°C, and fed to lower absorption section 3 of absorption tower 2.

The remainder (200 m³/hr) of partially regenerated absorbing fluid 21 was fed to a low-pressure regeneration tower 11 where it was depressurized to 0.85 kgG/cm² and regenerated with carbon dioxide and water discharged from the top of the tower. The bottom fluid was heated to about 110°C by means of a reboiler 12 provided at the lower part of low-pressure regeneration tower 11, and recycled to low-pressure regeneration tower 11. Thus, there was obtained a regenerated absorbing fluid 22 containing 5 Nm³ of carbon dioxide (CO₂) per m³ of the absorbing fluid. The liberated carbon dioxide and water, together with a small amount of the absorbing fluid, were cooled in a condenser 13 and introduced into a gas-liquid separator 14 where carbon dioxide 19 was separated. The cooled and separated carbon dioxide 19 had a temperature of about 40°C, a pressure of 0.35 kgG/cm² and a flow rate of 5,500 Nm³/hr.

The regenerated absorbing fluid 22 withdrawn from the bottom of low-pressure regeneration tower 11 had a

temperature of about 110°C and was fed to upper absorption section 4 of absorption tower 2. During this course, regenerated absorbing fluid 22 having a temperature of 110°C was subjected to heat exchange with loaded absorbing fluid 20 having a lower temperature, and further cooled to 38°C.

Of all the carbon dioxide recovered in the above-described manner, 90% was recovered in high-pressure regeneration tower 8 and 10% was recovered in low-pressure regeneration tower 11.

Thus, the carbon dioxide concentration of the raw gas for use in ammonia synthesis was reduced to a level sufficient for supply to an ammonia synthesis process. Moreover, the pressure of the recovered carbon dioxide was so high that the compression power required for the synthesis of urea could be saved and the scale of the equipment could be reduced.

Example 3

This example relates to an application of the process illustrated in the flow diagram of FIG. 1' in which natural gas was treated with an absorbing fluid comprising an aqueous solution containing 45% by weight of MDEA to remove carbon dioxide therefrom.

Natural gas 1 having a carbon dioxide concentration of 26% by volume was fed to the lower part of an absorption tower 2 under conditions including a pressure of 58 kgG/cm² and a temperature of 25°C.

A lower absorption section 3 of absorption tower 2 was packed, for example, with a wetted-wall type packing material, so that the ascending natural gas 1 came into efficient gas-liquid contact with a partially regenerated absorbing fluid 21. Thus, in lower absorption section 3, carbon dioxide was partially removed from natural gas 1 until its carbon dioxide concentration was reduced to about 3% by volume. Moreover, the ascending natural gas 1 came into efficient gas-liquid contact with a regenerated absorbing fluid 22 in an upper absorption section 4 comprising a similar wetted-wall type packed bed. The refined gas 17 was discharged from the top of absorption tower 2. This refined gas 17 had a carbon dioxide concentration of 50 ppm, a temperature of 50°C and a pressure of 58 kgG/cm².

The loaded absorbing fluid 20 having carbon dioxide absorbed therein was subjected to heat exchange, then heated to 130°C, and fed to a high-pressure regeneration tower 8 where it was partially regenerated.

In high-pressure regeneration tower 8, the carbon dioxide liberated by the aforesaid heating was separated from absorbing fluid 20 to obtain a partially regenerated absorbing fluid 21. Accordingly, the use of a heater such as a reboiler 12 is unnecessary, but such a heater may be used as required. The liberated carbon dioxide and water, together with a small amount of the absorbing fluid, were

cooled in a condenser 9 and introduced into a gas-liquid separator 10 where carbon dioxide 18 was separated. The cooled and separated carbon dioxide 18 had a temperature of about 40°C and a pressure of 55 kgG/cm² which was almost equal to the pressure of natural gas 1 used as the raw gas. This carbon dioxide 18 may be pressurized to 150 kgG/cm² by means of a compressor (not shown) and used for the purpose of tertiary oil recovery or storage in the earth.

On the other hand, the partially regenerated absorbing fluid 21 withdrawn from the bottom of high-pressure regeneration tower 8 had a temperature of about 130°C, and a predetermined portion thereof was fed to lower absorption section 3 of absorption tower 2. During this course, partially regenerated absorbing fluid 21 was subjected to heat exchange with loaded absorbing fluid 20 having a lower temperature, and further cooled to a required temperature.

The remainder of partially regenerated absorbing fluid 21 was fed to a low-pressure regeneration tower 11 where it was depressurized to 0.85 kgG/cm² and regenerated with carbon dioxide 19 and water discharged from the top of the tower. The bottom fluid was heated to about 130°C by means of a reboiler 12 provided at the lower part of low-pressure regeneration tower 11, and recycled to low-pressure regeneration tower 11. Thus, there was obtained a regenerated absorbing fluid 22 containing about 45% by weight

of MDEA. The liberated carbon dioxide and water, together with a small amount of the absorbing fluid, were cooled in a condenser 13 and introduced into a gas-liquid separator 14 where carbon dioxide 19 was separated. The cooled and
5 separated carbon dioxide 19 had a temperature of about 40°C and a pressure of 0.35 kgG/cm², and was used as an industrial gas.

The regenerated absorbing fluid 22 withdrawn from the bottom of low-pressure regeneration tower 11 had a
10 temperature of about 130°C and was fed to upper absorption section 4 of absorption tower 2. During this course, regenerated absorbing fluid 22 having a temperature of 130°C was subjected to heat exchange with loaded absorbing fluid 20 having a lower temperature, and further cooled to a required
15 temperature.

Of all the carbon dioxide recovered in the above-described manner, 85% was recovered in high-pressure regeneration tower 8 and 15% was recovered in low-pressure regeneration tower
11.

20 Thus, the carbon dioxide concentration of the refined natural gas 17 was reduced to a level which would not cause solid dry ice to be produced during the manufacture of LNG. Moreover, the pressure of the recovered carbon dioxide was so high that the compression power required for the production
25 of liquid carbon dioxide for use in tertiary oil recovery

could be saved and the scale of the equipment could be reduced.

Example 4

5 This example relates to an application of the process illustrated in the flow diagram of FIG. 1 in which raw hydrogen gas for use in chemical syntheses obtained by the reforming of natural gas was treated with an absorbing fluid comprising an aqueous solution containing 40% by weight of triethanolamine (TEA) and 4% by weight of piperazine as a reaction accelerator to remove carbon dioxide therefrom.

10 A raw gas 1 comprising 68 mole % of hydrogen, 12 mole % of methane, 19 mole % of carbon dioxide, and other inert gases was fed to the lower part of an absorption tower 2 under conditions including a pressure of 33 kgG/cm² and a temperature of 60°C.

15 In a lower absorption section 3 of absorption tower 2, the ascending raw gas 1 came into gas-liquid contact with a partially regenerated absorbing fluid 21. Thus, in lower absorption section 3, carbon dioxide was partially removed from raw gas 1 until its carbon dioxide concentration was reduced to about 1% by volume. Moreover, the ascending raw gas 1 came into gas-liquid contact with a regenerated absorbing fluid 22 in an upper absorption section 4. The refined gas 17, which had a carbon dioxide concentration of 200 ppm, a temperature of 40°C and a pressure of 32 kgG/cm²,

20

25

was discharged from the top of absorption tower 2.

The loaded absorbing fluid 20 having carbon dioxide absorbed thereinto was subjected to heat exchange, then heated to 120°C, and fed to a high-pressure regeneration tower 8 where about 60% of the carbon dioxide present in loaded absorbing fluid 20 was liberated. The liberated carbon dioxide and water, together with a small amount of the absorbing fluid, were cooled in a condenser 9 and introduced into a gas-liquid separator 10 where carbon dioxide 18 was separated. The cooled and separated carbon dioxide 18 had a temperature of about 40°C and a pressure of 9 kgG/cm². This carbon dioxide 18 was pressurized to 100 kgG/cm² by means of a compressor (not shown) and used for the production of liquid carbon dioxide.

On the other hand, the partially regenerated absorbing fluid 21 withdrawn from the bottom of high-pressure regeneration tower 8 was subjected to heat exchange with loaded absorbing fluid 20, further cooled, and fed to lower absorption section 3 of absorption tower 2.

The remainder (about 10%) of partially regenerated absorbing fluid 21 was fed to a low-pressure regeneration tower 11 where it was depressurized to 0.85 kgG/cm² and regenerated with carbon dioxide 19 and water discharged from the top of the tower. The bottom fluid was heated to about 130°C by means of a reboiler 12 provided at the lower part of

low-pressure regeneration tower 11, and recycled to low-pressure regeneration tower 11. From the resulting regenerated absorbing fluid 22, about 98% of the carbon dioxide present in the starting loaded absorbing fluid 20 had been liberated. The carbon dioxide 19 separated by means of a gas-liquid separator 14 had a temperature of about 40°C and a pressure of 0.35 kgG/cm². This carbon dioxide 19 was also pressurized to 100 kgG/cm² by means of a compressor and used for the production of liquid carbon dioxide.

The regenerated absorbing fluid 22 withdrawn from the bottom of low-pressure regeneration tower 11 had a temperature of about 130°C and was fed to upper absorption section 4 of absorption tower 2. During this course, regenerated absorbing fluid 22 was subjected to heat exchange with loaded absorbing fluid 20 having a lower temperature, and further cooled with cooling water.

Of all the carbon dioxide recovered in the above-described manner, 92% was recovered in high-pressure regeneration tower 8 and 8% was recovered in low-pressure regeneration tower 11.

Thus, the carbon dioxide concentration of the raw hydrogen gas for use in chemical syntheses was reduced to a level sufficient for supply to subsequent process steps. Moreover, the proportion of the recovered high-pressure carbon dioxide was so high that the compression power required for the production of liquid carbon dioxide could be saved and the

scale of the equipment could be reduced.

Comparative Example 1

Using the same high-pressure natural gas and absorbing fluid as used in Example 3, the natural gas was treated in a conventional process (see FIG. 2) in order to reduce its carbon dioxide concentration to 50 ppm and, moreover, recover carbon dioxide.

The carbon dioxide-loaded absorbing fluid 120 was fed to a first flash drum 123 where it was flashed under a pressure of 3.8 kgG/cm² to liberate a portion of the carbon dioxide present in loaded absorbing fluid 120 and thereby obtain a partially regenerated absorbing fluid. This partially regenerated absorbing fluid is further heated in a heater 107 and fed to a second flash drum 124 where it was flashed again to obtain a regenerated absorbing fluid 122.

The carbon dioxide obtained in first flash drum 123 had a pressure of 3.8 kgG/cm², which was far lower than the pressure (55 kgG/cm²) of the carbon dioxide obtained in Example 1. Accordingly, in order to produce liquid carbon dioxide for use in tertiary oil recovery, the process of Comparative Example 1 was more disadvantageous than the process of Example 1 from the viewpoint of compression power and equipment. In this process, natural gas 101 is fed to absorption tower 102 and discharged as a refined gas 117.

Numerical 109 designates an overhead condenser disposed between

second flash drum 124 and separating drum 110, numeral 118 designates the carbon dioxide discharged from separating drum 110, numeral 132 designates the carbon dioxide discharged from first flash drum 123, and numeral 116 designates a cooler.

5

2
3
4
5
6
7
8
9
10
11
12
13
14
15
16
17
18
19
20
21
22
23
24
25
26
27
28
29
30
31
32
33
34
35
36
37
38
39
40
41
42
43
44
45
46
47
48
49
50
51
52
53
54
55
56
57
58
59
60
61
62
63
64
65
66
67
68
69
70
71
72
73
74
75
76
77
78
79
80
81
82
83
84
85
86
87
88
89
90
91
92
93
94
95
96
97
98
99
100
101
102
103
104
105
106
107
108
109
110
111
112
113
114
115
116
117
118
119
120
121
122
123
124
125
126
127
128
129
130
131
132
133
134
135
136
137
138
139
140
141
142
143
144
145
146
147
148
149
150
151
152
153
154
155
156
157
158
159
160
161
162
163
164
165
166
167
168
169
170
171
172
173
174
175
176
177
178
179
180
181
182
183
184
185
186
187
188
189
190
191
192
193
194
195
196
197
198
199
200
201
202
203
204
205
206
207
208
209
210
211
212
213
214
215
216
217
218
219
220
221
222
223
224
225
226
227
228
229
230
231
232
233
234
235
236
237
238
239
240
241
242
243
244
245
246
247
248
249
250
251
252
253
254
255
256
257
258
259
260
261
262
263
264
265
266
267
268
269
270
271
272
273
274
275
276
277
278
279
280
281
282
283
284
285
286
287
288
289
290
291
292
293
294
295
296
297
298
299
300
301
302
303
304
305
306
307
308
309
310
311
312
313
314
315
316
317
318
319
320
321
322
323
324
325
326
327
328
329
330
331
332
333
334
335
336
337
338
339
340
341
342
343
344
345
346
347
348
349
350
351
352
353
354
355
356
357
358
359
360
361
362
363
364
365
366
367
368
369
370
371
372
373
374
375
376
377
378
379
380
381
382
383
384
385
386
387
388
389
390
391
392
393
394
395
396
397
398
399
400
401
402
403
404
405
406
407
408
409
410
411
412
413
414
415
416
417
418
419
420
421
422
423
424
425
426
427
428
429
430
431
432
433
434
435
436
437
438
439
440
441
442
443
444
445
446
447
448
449
450
451
452
453
454
455
456
457
458
459
460
461
462
463
464
465
466
467
468
469
470
471
472
473
474
475
476
477
478
479
480
481
482
483
484
485
486
487
488
489
490
491
492
493
494
495
496
497
498
499
500
501
502
503
504
505
506
507
508
509
510
511
512
513
514
515
516
517
518
519
520
521
522
523
524
525
526
527
528
529
530
531
532
533
534
535
536
537
538
539
540
541
542
543
544
545
546
547
548
549
550
551
552
553
554
555
556
557
558
559
560
561
562
563
564
565
566
567
568
569
570
571
572
573
574
575
576
577
578
579
580
581
582
583
584
585
586
587
588
589
590
591
592
593
594
595
596
597
598
599
600
601
602
603
604
605
606
607
608
609
610
611
612
613
614
615
616
617
618
619
620
621
622
623
624
625
626
627
628
629
630
631
632
633
634
635
636
637
638
639
640
641
642
643
644
645
646
647
648
649
650
651
652
653
654
655
656
657
658
659
660
661
662
663
664
665
666
667
668
669
670
671
672
673
674
675
676
677
678
679
680
681
682
683
684
685
686
687
688
689
690
691
692
693
694
695
696
697
698
699
700
701
702
703
704
705
706
707
708
709
710
711
712
713
714
715
716
717
718
719
720
721
722
723
724
725
726
727
728
729
730
731
732
733
734
735
736
737
738
739
740
741
742
743
744
745
746
747
748
749
750
751
752
753
754
755
756
757
758
759
760
761
762
763
764
765
766
767
768
769
770
771
772
773
774
775
776
777
778
779
780
781
782
783
784
785
786
787
788
789
790
791
792
793
794
795
796
797
798
799
800
801
802
803
804
805
806
807
808
809
810
811
812
813
814
815
816
817
818
819
820
821
822
823
824
825
826
827
828
829
830
831
832
833
834
835
836
837
838
839
840
841
842
843
844
845
846
847
848
849
850
851
852
853
854
855
856
857
858
859
860
861
862
863
864
865
866
867
868
869
870
871
872
873
874
875
876
877
878
879
880
881
882
883
884
885
886
887
888
889
890
891
892
893
894
895
896
897
898
899
900
901
902
903
904
905
906
907
908
909
910
911
912
913
914
915
916
917
918
919
920
921
922
923
924
925
926
927
928
929
930
931
932
933
934
935
936
937
938
939
940
941
942
943
944
945
946
947
948
949
950
951
952
953
954
955
956
957
958
959
960
961
962
963
964
965
966
967
968
969
970
971
972
973
974
975
976
977
978
979
980
981
982
983
984
985
986
987
988
989
990
991
992
993
994
995
996
997
998
999
1000
1001
1002
1003
1004
1005
1006
1007
1008
1009
1010
1011
1012
1013
1014
1015
1016
1017
1018
1019
1020
1021
1022
1023
1024
1025
1026
1027
1028
1029
1030
1031
1032
1033
1034
1035
1036
1037
1038
1039
1040
1041
1042
1043
1044
1045
1046
1047
1048
1049
1050
1051
1052
1053
1054
1055
1056
1057
1058
1059
1060
1061
1062
1063
1064
1065
1066
1067
1068
1069
1070
1071
1072
1073
1074
1075
1076
1077
1078
1079
1080
1081
1082
1083
1084
1085
1086
1087
1088
1089
1090
1091
1092
1093
1094
1095
1096
1097
1098
1099
1100
1101
1102
1103
1104
1105
1106
1107
1108
1109
1110
1111
1112
1113
1114
1115
1116
1117
1118
1119
1120
1121
1122
1123
1124
1125
1126
1127
1128
1129
1130
1131
1132
1133
1134
1135
1136
1137
1138
1139
1140
1141
1142
1143
1144
1145
1146
1147
1148
1149
1150
1151
1152
1153
1154
1155
1156
1157
1158
1159
1160
1161
1162
1163
1164
1165
1166
1167
1168
1169
1170
1171
1172
1173
1174
1175
1176
1177
1178
1179
1180
1181
1182
1183
1184
1185
1186
1187
1188
1189
1190
1191
1192
1193
1194
1195
1196
1197
1198
1199
1200
1201
1202
1203
1204
1205
1206
1207
1208
1209
1210
1211
1212
1213
1214
1215
1216
1217
1218
1219
1220
1221
1222
1223
1224
1225
1226
1227
1228
1229
1230
1231
1232
1233
1234
1235
1236
1237
1238
1239
1240
1241
1242
1243
1244
1245
1246
1247
1248
1249
1250
1251
1252
1253
1254
1255
1256
1257
1258
1259
1260
1261
1262
1263
1264
1265
1266
1267
1268
1269
1270
1271
1272
1273
1274
1275
1276
1277
1278
1279
1280
1281
1282
1283
1284
1285
1286
1287
1288
1289
1290
1291
1292
1293
1294
1295
1296
1297
1298
1299
1300
1301
1302
1303
1304
1305
1306
1307
1308
1309
1310
1311
1312
1313
1314
1315
1316
1317
1318
1319
1320
1321
1322
1323
1324
1325
1326
1327
1328
1329
1330
1331
1332
1333
1334
1335
1336
1337
1338
1339
1340
1341
1342
1343
1344
1345
1346
1347
1348
1349
1350
1351
1352
1353
1354
1355
1356
1357
1358
1359
1360
1361
1362
1363
1364
1365
1366
1367
1368
1369
1370
1371
1372
1373
1374
1375
1376
1377
1378
1379
1380
1381
1382
1383
1384
1385
1386
1387
1388
1389
1390
1391
1392
1393
1394
1395
1396
1397
1398
1399
1400
1401
1402
1403
1404
1405
1406
1407
1408
1409
1410
1411
1412
1413
1414
1415
1416
1417
1418
1419
1420
1421
1422
1423
1424
1425
1426
1427
1428
1429
1430
1431
1432
1433
1434
1435
1436
1437
1438
1439
1440
1441
1442
1443
1444
1445
1446
1447
1448
1449
1450
1451
1452
1453
1454
1455
1456
1457
1458
1459
1460
1461
1462
1463
1464
1465
1466
1467
1468
1469
1470
1471
1472
1473
1474
1475
1476
1477
1478
1479
1480
1481
1482
1483
1484
1485
1486
1487
1488
1489
1490
1491
1492
1493
1494
1495
1496
1497
1498
1499
1500
1501
1502
1503
1504
1505
1506
1507
1508
1509
1510
1511
1512
1513
1514
1515
1516
1517
1518
1519
1520
1521
1522
1523
1524
1525
1526
1527
1528
1529
1530
1531
1532
1533
1534
1535
1536
1537
1538
1539
1540
1541
1542
1543
1544
1545
1546
1547
1548
1549
1550
1551
1552
1553
1554
1555
1556
1557
1558
1559
1560
1561
1562
1563
1564
1565
1566
1567
1568
1569
1570
1571
1572
1573
1574
1575
1576
1577
1578
1579
1580
1581
1582
1583
1584
1585
1586
1587
1588
1589
1590
1591
1592
1593
1594
1595
1596
1597
1598
1599
1600
1601
1602
1603
1604
1605
1606
1607
1608
1609
1610
1611
1612
1613
1614
1615
1616
1617
1618
1619
1620
1621
1622
1623
1624
1625
1626
1627
1628
1629
1630
1631
1632
1633
1634
1635
1636
1637
1638
1639
1640
1641
1642
1643
1644
1645
1646
1647
1648
1649
1650
1651
1652
1653
1654
1655
1656
1657
1658
1659
1660
1661
1662
1663
1664
1665
1666
1667
1668
1669
1670
1671
1672
1673
1674
1675
1676
1677
1678
1679
1680
1681
1682
1683
1684
1685
1686
1687
1688
1689
1690
1691
1692
1693
1694
1695
1696
1697
1698
1699
1700
1701
1702
1703
1704
1705
1706
1707
1708
1709
1710
1711
1712
1713
1714
1715
1716
1717
1718
1719
1720
1721
1722
1723
1724
1725
1726
1727
1728
1729
1730
1731
1732
1733
1734
1735
1736
1737
1738
1739
1740
1741
1742
1743
1744
1745
1746
1747
1748
1749
1750
1751
1752
1753
1754
1755
1756
1757
1758
1759
1760
1761
1762
1763
1764
1765
1766
1767
1768
1769
1770
1771
1772
1773
1774
1775
1776
1777
1778
1779
1780
1781
1782
1783
1784
1785
1786
1787
1788
1789
1790
1791
1792
1793
1794
1795
1796
1797
1798
1799
1800
1801
1802
1803
1804
1805
1806
1807
1808
1809
1810
1811
1812
1813
1814
1815
1816
1817
1818
1819
1820
1821
1822
1823
1824
1825
1826
1827
1828
1829
1830
1831
1832
1833
1834
1835
1836
1837
1838
1839
1840
1841
1842
1843
1844
1845
1846
1847
1848
1849
1850
1851
1852
1853
1854
1855
1856
1857
1858
1859
1860
1861
1862
1863
1864
1865
1866
1867
1868
1869
1870
1871
1872
1873
1874
1875
1876
1877
1878
1879
1880
1881
1882
1883
1884
1885
1886
1887
1888
1889
1890
1891
1892
1893
1894
1895
1896
1897
1898
1899
1900
1901
1902
1903
1904
1905
1906
1907
1908
1909
1910
1911
1912
1913
1914
1915
1916
1917
1918
1919
1920
1921
1922
1923
1924
1925
1926
1927
1928
1929
1930
1931
1932
1933
1934
1935
1936
1937
1938
1939
1940
1941
1942
1943
1944
1945
1946
1947
1948
1949
1950
1951
1952
1953
1954
1955
1956
1957
1958
1959
1960
1961
1962
1963
1964
1965
1966
1967
1968
1969
1970
1971
1972
1973
1974
1975
1976
1977
1978
1979
1980
1981
1982
1983
1984
1985
1986
1987
1988
1989
1990
1991
1992
1993
1994
1995
1996
1997
1998
1999
2000
2001
2002
2003
2004
2005
2006
2007
2008
2009
2010
2011
2012
2013
2014
2015
2016
2017
2018
2019
2020
2021
2022
2023
2024
2025
2026
2027
2028
2029
2030
2031
2032
2033
2034
2035
2036
2037
2038
2039
2040
2041
2042
2043
2044
2045
2046
2047
2048
2049
2050
2051
2052
2053
2054
2055
2056
2057
2058
2059
2060
2061
2062
2063
2064
2065
2066
2067
2068
2069
2070
2071
2072
2073
2074
2075
2076
2077
2078
2079
2080
2081
2082
2083
2084
2085
2086
2087
2088
2089
2090
2091
2092
2093
2094
2095
2096
2097
2098
2099
2100
2101
2102
2103
2104
2105
2106
2107
2108
2109
2110
2111
2112
2113
2114
2115
2116
2117
2118
2119
2120
2121
2122
2123
2124
2125
2126
2127
2128
2129
2130
2131
2132
2133
2134
2135
2136
2137
2138
2139
2140
2141
2142
2143
2144
2145
2146
2147
2148
2149
2150
2151
2152
2153
2154
2155
2156
2157
2158
2159
2160
2161
2162
2163
2164
2165
2166
2167
2168
2169
2170
2171
2172
2173
2174
2175
2176
2177
2178
2179
2180
2181
2182
2183
2184
2185
2186
2187
2188
2189
2190
2191
2192
2193
2194
2195
2196
2197
2198
2199
2200
2201
2202
2203
2204
2205
2206
2207
2208
2209
2210
2211
2212
2213
2214
2215
2216
2217
2218
2219
2220
2221
2222
2223
2

The claims defining the invention are as follows:-

1. A process for the removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas which comprises the steps of feeding a high-pressure raw gas
5 containing carbon dioxide to a decarbonation tower having a lower absorption section and an upper absorption section, where the raw gas is brought into gas-liquid contact with a partially regenerated absorbing fluid in the lower absorption section so as to cause carbon dioxide to be partially
10 absorbed thereinto, and then brought into gas-liquid contact with a regenerated absorbing fluid in the upper absorption section so as to cause carbon dioxide to be absorbed thereinto until a very low carbon dioxide concentration is reached, and the carbon dioxide-free refined gas is
15 discharged out of the system; heating the carbon dioxide-loaded absorbing fluid produced in the decarbonation tower and feeding it to a high-pressure regeneration tower where some carbon dioxide is liberated under pressure to obtain a partially regenerated absorbing fluid, and a portion of the
20 partially regenerated absorbing fluid is fed to the lower absorption section; feeding the remainder of the partially regenerated absorbing fluid to a low-pressure regeneration tower where carbon dioxide is liberated to obtain a regenerated absorbing fluid, and the regenerated absorbing
25 fluid is fed to the upper absorption section; recovering

high-pressure carbon dioxide by cooling the carbon dioxide liberated under pressure in the high-pressure regeneration tower and separating it from any entrained water; and recovering carbon dioxide by cooling the carbon dioxide liberated in the low-pressure regeneration tower and separating it from any entrained water.

2. A process for the removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas according to claim 1 wherein the high-pressure raw gas has a carbon dioxide concentration of 2 to 50%.

3. A process for the removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas according to claim 1 or 2 wherein the high-pressure raw gas has a pressure of not less than 2 kg/cm² (absolute pressure).

4. A process for the removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas according to any one of claims 1-3 wherein the refined gas has a carbon dioxide concentration of 10 to 10,000 ppm.

5. A process for the removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas according to any one of claims 1-4 wherein the heating temperature of the carbon dioxide-loaded absorbing fluid at which some carbon dioxide is liberated from the carbon dioxide-loaded absorbing fluid to obtain a partially regenerated absorbing fluid is in the range of 90 to 150°C.

6. A process for the removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas according to any one of claims 1-5 wherein the pressure under which some carbon dioxide is liberated from the carbon dioxide-loaded absorbing fluid to obtain a partially regenerated absorbing fluid ranges from 2 kg/cm² (absolute pressure) to the pressure of the raw gas.

7. A process for the removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas according to any one of claims 1-6 wherein the heating temperature of the partially regenerated absorbing fluid at which carbon dioxide is liberated from the remainder of the partially regenerated absorbing fluid to obtain a regenerated absorbing fluid is in the range of 100 to 150°C.

8. A process for the removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas according to any one of claims 1-7 wherein the pressure under which carbon dioxide is liberated from the remainder of the partially regenerated absorbing fluid to obtain a regenerated absorbing fluid is less than 2 kg/cm² (absolute pressure).

9. A system for the removal and high-pressure recovery of carbon dioxide from a high-pressure raw gas which comprises a decarbonation tower having a lower absorption section and an upper absorption section, a heater for a carbon dioxide-loaded absorbing fluid, a high-pressure regeneration tower

for the carbon dioxide-loaded absorbing fluid, a cooler and a gas-liquid separator provided at the top of the high-pressure regeneration tower, a low-pressure regeneration tower for a partially regenerated absorbing fluid, a heater provided at the bottom of the low-pressure regeneration tower, and a cooler and a gas-liquid separator provided at the top of the low-pressure regeneration tower, whereby a high-pressure raw gas containing carbon dioxide is fed to the decarbonation tower where the raw gas is brought into gas-liquid contact with the partially regenerated absorbing fluid in the lower absorption section so as to cause carbon dioxide to be partially absorbed thereinto, and then brought into gas-liquid contact with the regenerated absorbing fluid in the upper absorption section so as to cause carbon dioxide to be absorbed thereinto until a very low carbon dioxide concentration is reached, and the carbon dioxide-free refined gas is discharged out of the system; the carbon dioxide-loaded absorbing fluid produced in the decarbonation tower is heated and fed to the high-pressure regeneration tower where some carbon dioxide is liberated under pressure to obtain a partially regenerated absorbing fluid, and a portion of the partially regenerated absorbing fluid is fed to the lower absorption section; the remainder of the partially regenerated absorbing fluid is fed to a low-pressure regeneration tower where carbon dioxide is liberated to

obtain a regenerated absorbing fluid, and the regenerated absorbing fluid is fed to the upper absorption section; high-pressure carbon dioxide is recovered by cooling the carbon dioxide liberated under pressure in the high-pressure regeneration tower and separating it from any entrained water by means of the cooler and gas-liquid separator provided at the top of the high-pressure regeneration tower; and carbon dioxide is recovered by cooling the carbon dioxide liberated in the low-pressure regeneration tower and separating it from any entrained water by means of the cooler and gas-liquid separator provided at the top of the low-pressure regeneration tower.

10. A process for the removal and high-pressure recoveries of carbon dioxide from a high-pressure raw gas substantially as hereinbefore described in any one of Examples 1 to 4.

11. A system for the removal and high-pressure recoveries of carbon dioxide from a high-pressure raw gas substantially as hereinbefore described with reference to Figure 1 of the accompanying drawings.

DATED THIS 10TH DAY OF JULY 1997

10
MITSUBISHI HEAVY INDUSTRIES, LTD
BY THEIR PATENT ATTORNEYS
LORD & COMPANY
PERTH, WESTERN AUSTRALIA

FIG. 1

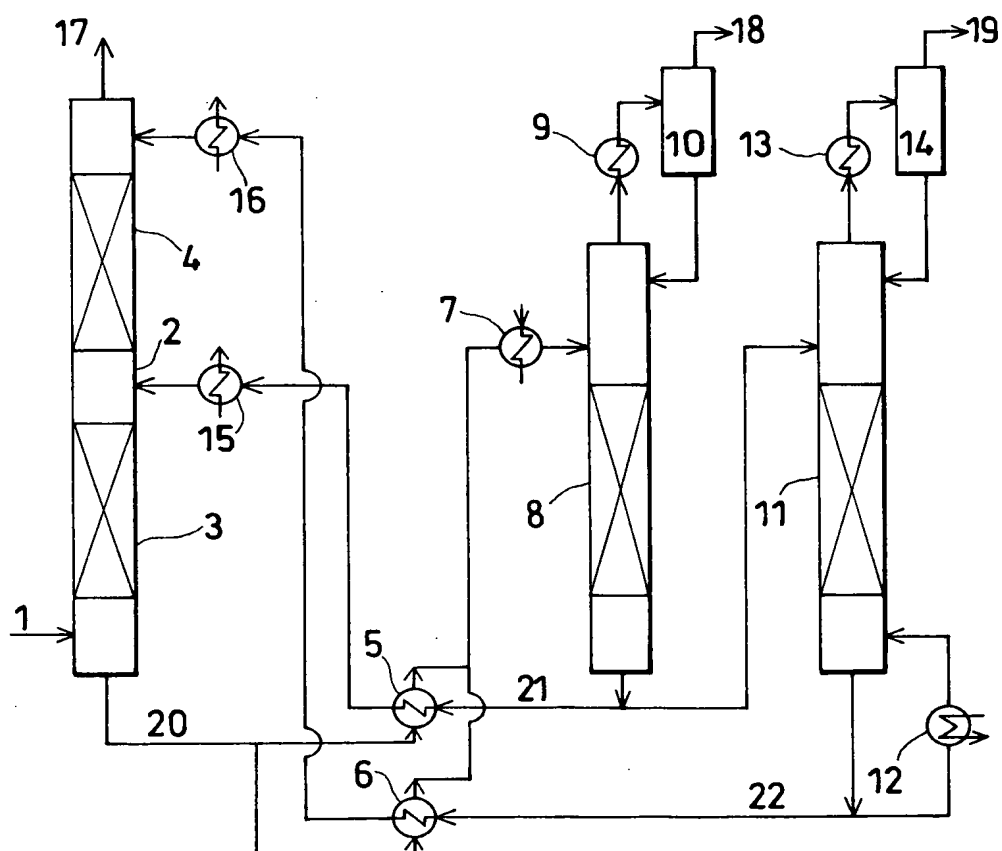


FIG. 2

